Response to Reviewers' comments on "Microphysics-based black carbon aging in a global CTM: constraints from HIPPO observations and implications for global black carbon budget" by He et al.

Referee #1

"… This paper represents an important step towards a more process-oriented treatment of BC aging in global models. The analysis is of great interest to the community and the paper fits well into the scope of ACP. A weakness of this paper is the mathematical description of the parameterization and the underlying assumptions. I recommend the paper for publication after the following questions and comments are addressed."

Response: We thank the reviewer for constructive comments and suggestions. We have provided itemized responses to the following specific comments. Particularly, we have improved the mathematical descriptions of the parameterization and underlying assumptions.

Major Comments:

1.1 *"Before starting with section 2.2.1, please describe briefly the aerosol scheme used in GEOS-Chem more clearly – what are the prognostic variables? From the text that follows it sounds like this is a bulk scheme, and only mass concentrations of different aerosol species are tracked, but it would be useful to mention this early on."*

Response: Points are well taken. We have included additional descriptions of the GEOS-Chem aerosol scheme in Pages 8-9, Lines 178-193 (in the track-change manuscript) as follows:

"GEOS-Chem includes a fully coupled treatment of tropospheric O_3-NO_x-VOC chemistry, sulfate-nitrate-ammonia and carbonaceous aerosols. Park et al. (2003) presented the first GEOS-Chem simulation of carbonaceous aerosols including BC and organic carbon (OC). The model also accounts for other aerosols including secondary organic aerosol (SOA), dust, and sea salt. GEOS-Chem uses a bulk aerosol scheme that separately tracks mass concentrations of different aerosol species (i.e., externally mixed). The model resolves hydrophobic and hydrophilic BC and OC, fine-mode (0.01–0.5 μm) and coarse-mode (0.5–8.0 μm) sea salt, dust in four size bins (0.1–1.0, 1.0–1.8, 1.8–3.0, and 3.0–6.0 μm), and five types of lumped SOA formed from different precursors. Aerosol and gas phase simulations are coupled through formation of sulfate, nitrate, and SOA, heterogeneous chemistry, and aerosol effects on photolysis rates. Details on the GEOS-Chem aerosol simulations are provided, respectively, by Park et al. (2003) for BC and OC, Park et al. (2004) for sulfate-nitrate-ammonia, Liao et al. (2007) for SOA, Fairlie et al. (2007) for dust, and Alexander et al. (2005) for sea salt."

1.2 *“Equation (3): What expression for the correction factor f is used? What is the value for α?”*

Response: We have included the expression (Dahneke, 1983) for the correction factor in Eq. (3) as follows:
“

$$f(K_n,\alpha)=\frac{1+K_n}{1+2K_n(1+K_n)/\alpha} \qquad (4)”$$

In this study, α = 1. We have added the corresponding statement in Page 10, Lines 218-221 as follows:
“…, α is the accommodation coefficient (α = 1 in this study), and $f(K_n, \alpha)$ is the correction factor for non-continuum effects and imperfect surface accommodation based on the mathematical expression from Dahneke (1983).”

1.3 *“Equation (4): Please write out the limits for the integration and summation. How is the integral numerically evaluated?”*

Response: We have added the limits for the integration and summation as follows:
“

$$k_A=\frac{J_{A,BCPO,tot}}{\sum_{p_{i=1}}^{p_{i=7}} J_{A,p_i,tot}}M_{A,cond}=\frac{\int_0^{\infty} n_{BCPO}(R_{BCPO})\,f_{BCPO}(K_n,\alpha)R_{BCPO}dR_{BCPO}}{\sum_{p_{i=1}}^{p_{i=7}}\int_0^{\infty} n_{p_i}(R_{p_i})\,f_{p_i}(K_n,\alpha)R_{p_i}dR_{p_i}}M_{A,cond} \qquad (5)”$$

We have also included a description of this equation in Pages 10-11, Lines 228-233 as follows:
“where p_i (i = 1–7) represents seven types of pre-existing aerosols (i.e., BCPO, BCPI, hydrophobic OC, hydrophilic OC, sulfate, fine-mode and coarse-mode sea salt) available for condensation, $J_{A,p_i,tot}$ is the condensation rate of A onto particle p_i, $M_{A,cond}$ is the total condensed mass of A in a model grid per unit time, R_{p_i} and n_{p_i} (= dN_{p_i}/dR_{p_i}) are the radius and number concentration distribution function of pre-existing particles, respectively.”

For the computation of the integral, we assumed a constant correction factor $f_{p_i}(K_n,\alpha)$ for each type of aerosols with different sizes. Thus, this factor can be moved out of the integral for each aerosol type. By assuming a lognormal aerosol size distribution (see Eq. (6) in the track-changed manuscript), the integral in Eq. (5) can be computed analytically by using the following mathematical identity:

$$\int_0^\infty n(R)\cdot RdR = N_{tot}\frac{D_g}{2}\exp(\frac{1}{2}\ln^2\sigma_g)$$

where N_{tot} is the total particle number concentration, D_g is the geometric mean diameter, and σ_g is the geometric standard deviation. We realize that assuming a constant correction factor for each aerosol species with different sizes may introduce uncertainty into our calculations. We have included these discussions in Pages 11-12, Lines 250-255 as follows:

"In order to analytically compute the integral in Eq. (5), we have assumed a constant correction factor $f(K_n, \alpha)$ for each type of aerosols with different sizes, which may introduce uncertainty in the computation. Under this assumption and using a lognormal aerosol size distribution, the integral can now be computed by following the mathematical identity:

$$\int_0^\infty n_{p_i}(R_{p_i})R_{p_i}dR_{p_i} = N_{p_i}\frac{D_{p_i}}{2}\exp(\frac{1}{2}\ln^2\sigma_{p_i}) \qquad (7)$$"

1.4 *"Page 32786, line 23: Please rephrase the explanation for subscript i (it clearly doesn't represent various pre-existing particles. Do you mean sub-populations or classes?"*

Response: We thank the reviewer for pointing this out. To clarify, we have changed "i" to "p_i" in Eq. (5), which represents different types of pre-existing aerosols (i.e., BCPO, BCPI, hydrophobic OC, hydrophilic OC, sulfate, fine-mode and coarse-mode sea salt) available for condensation. Moreover, we did not consider sub-populations or sub-classes for each type of aerosols in this study. We have included the corresponding clarification in Pages 10-11, Lines 228-233 (please see the response to Reviewer #1, comment 1.3).

1.5 *"Equation (4) and (5): Please distinguish between the number concentration and the number concentration distribution function (dN/dDp). In equation (4) it should be the latter. Usually a lower case n is used as variable for this."*

Response: Points are well taken. We have changed the notation "N" to "n" in Eq. (5) to represent the number concentration distribution function (dN/dR), while the "N_p" in Eq. (6) denotes the number concentration. We have included the corresponding clarification in Pages 10-11, Lines 228-245 (please see the response to Reviewer #1, comments 1.3 and 1.6).

1.6 *"Equation (5): Np should also have an index i (for the different classes). What are the values for the particles density?"*

Response: To clarify and to be consistent throughout the entire text, we have changed "N_p" to "N_{p_i}" in the equation, where p_i (i = 1–7) represents seven types of pre-existing aerosols (i.e., BCPO, BCPI, hydrophobic OC, hydrophilic OC, sulfate, fine-mode and coarse-mode sea salt) available for condensation (see also the response to Reviewer #1, comments 1.3 and 1.4). We did not consider different sub-classes for each type of aerosols in this study. In addition, we have added values for the density of different particles in Page 11, Lines 238-245 as follows:

"We convert aerosol mass concentration (m_{p_i}) to number concentration (N_{p_i}), assuming lognormal distributions for different aerosols following Croft et al. (2005) in the form:

$$N_{p_i} = \frac{m_{p_i}}{\rho_{p_i}} \left(\frac{\pi}{6} D_{p_i}^3 \exp\left(\frac{9}{2} \ln^2 \sigma_{p_i}\right)\right)^{-1} \tag{6}$$

where ρ_{p_i} is the particle density (1.8 g cm^{-3} for BC and OC, 1.7 g cm^{-3} for sulfate, and 2.2 g cm^{-3} for sea salt), D_{p_i} and σ_{p_i} are the geometric mean diameter and standard deviation of number size distribution, respectively."

1.7 *"Equation (7): Please write out the limits of this summation. Also, this is assuming that the different secondary aerosol species have the same hygroscopicity. Please state this assumption. SOA should actually be less hygroscopic than for example sulfuric acid and ammonium nitrate."*

Response: Points are well taken. We have added the limits of this summation and the corresponding descriptions and assumptions in Page 12, Lines 261-266 as follows:

"

$$F_{BCPO->BCPI} = \frac{\sum_{A_i=1}^{A_i=4} k_{A_i} \cdot \Delta t}{\beta M_{BCPO}} \tag{9}$$

where $F_{BCPO->BCPI}$ represents the fraction of BCPO becoming BCPI through condensation of four types of soluble species A_i (i = 1–4, i.e., H_2SO_4, HNO_3, NH_3, and SOA) in a model time step (Δt). This implicitly assumes that different secondary aerosol species have the same hygroscopicity. We note that some SOA species could be less hygroscopic than ammonia sulfate (Prenni et al., 2007)."

1.8 "*After shifting the hydrophobic mass of BC into the hydrophilic category, how do you treat the associated secondary aerosol material? Do you track it separately from the mass of sulfate/nitrate/ammonium/SOA that is not mixed with BC. (This comes back to comment 1.1 – what are the prognostic variables?"*

Response: We thank the reviewer for this comment. After converting hydrophobic BC to hydrophilic BC, we lump the associated SOA and sulfate-nitrate-ammonia with the mass of those not mixed with BC, respectively, because GEOS-Chem separately tracks mass concentrations of different aerosol species (see also the response to Reviewer #1, comment 1.1). We note that the lumping, instead of tracking the coated secondary aerosol material and hydrophilic BC together, may introduce relatively small uncertainty, because the size distribution (i.e., geometric mean diameter and standard deviation) of hydrophilic BC is similar to that of SOA and sulfate-nitrate-ammonia in this study (see Table 1). Furthermore, our sensitivity analysis (see Section 4 in text) showed that the size distributions of hydrophilic particles (including SOA, sulfate, and hydrophilic BC) only have minor effects on global BC distribution and lifetime. We have included the clarification in Pages 12-13, Lines 271-279 as follows:

"After the hydrophobic-to-hydrophilic BC conversion, we lump the mass of secondary aerosol materials coated on BC with those not mixed with BC in order to be compatible with the bulk aerosol scheme in GEOS-Chem, where the mass concentration of different aerosol species are separately tracked. The lumping, instead of treating coating materials and hydrophilic BC together, only introduces small uncertainty, considering that the size distribution of hydrophilic BC is similar to that of SOA and sulfate in this study (see Table 1). Further sensitivity analysis also show minor effects of the hydrophilic aerosol size distribution on global BC concentration and lifetime (see Sect. 4.3)."

1.9 *"Equation (9): This is the coagulation kernel that applies to the coagulation of two particles with radii* R_{BCPO} *and* R_X*. In the section for condensation you described that you are assuming a log-normal size distribution. How do you reconcile this with assuming specific radii in equation (9)?"*

Response: We thank the reviewer for pointing this out. In this study, we use geometric mean radii for R_{BCPO} and R_X. This may introduce uncertainty for particle sizes largely deviating from the mean value. However, we note that the resulting uncertainty in global BC distribution and lifetime could be small, because our results show that coagulation only contributes to a small part of total BC aging rate globally compared with condensation (see Sect. 3.1 in the text). Moreover, sensitivity analyses suggest that aerosol size distributions have minor impacts on global BC distribution (see Sects. 4.2 and 4.3 in the text). We have included these discussions in Page 13, Lines 293-298 as follows:

"We use geometric mean radii for R_{BCPO} and R_X, which could introduce uncertainty for particle sizes largely deviating from the mean value. We note that the resulting uncertainty in BC concentration and lifetime is likely small, because model results show that coagulation only makes a small contribution to the total BC aging rate over the globe (see Sect. 3.1) and the global BC distribution is insensitive to aerosol size distribution in this study (see Sects. 4.2 and 4.3).”

1.10 *“Equation (10): Please write out what the limits are for the summation and rephrase the explanation for index i.”*

Response: Points are well taken. We have added the limits for the summation and rephrased the statement of index in Page 14, Lines 302-305 as follows:
“

$$\left(\frac{dN_{BCPO}}{dt}\right)_{coag} = -\sum_{p_{j=1}}^{p_{j=6}} \gamma_{p_j} K_{BCPO,p_j} N_{p_j} N_{BCPO} \tag{12}$$

where p_j (j = 1–6) represents six types of hydrophilic particles, including sulfate-nitrate-ammonia, BCPI, hydrophilic OC, SOA, fine-mode and coarse-mode sea salt.”

1.11 *“What is the additional computational burden associated with the new parameterization compared to using the fixed aging time scale?”*

Response: Compared with the use of a fixed aging time, the computational burden associated with this new parameterization is caused by more calculations at each model time step as well as the requirement to run the fully coupled gas-aerosol chemistry instead of the uncoupled offline BC simulation only (the fixed aging scheme in GEOS-Chem does not include interaction between BC and other aerosols).

2. *“Setup of sensitivity studies: A potentially important sensitivity run that is missing is one that investigates the sensitivity to the assumption that 80”*

Response: This comment appears to be an incomplete sentence and as such we are unable to provide an appropriate response.

3. *“Given that there are several papers in the literature that use the aging time scale to present their results, it would be helpful to include a figure that shows a map of actual aging time scales and/or a pdf (similar to Figure 4, but instead of BC concentrations, show τ).”*

Response: Following the reviewer’s comment, we have included an additional figure to show the probability density function (PDF) of annual mean aging time over the globe. Please see the added Fig. 3 in the track-changed manuscript. We have also included corresponding discussions on this figure in Page 19, Lines 436-441 as follows:
“Figure 3 shows the probability density function (PDF) of simulated annual mean BC e-folding aging time (τ in Eq. 1) over the globe. The first PDF peak around τ = 8 h

represents the fast aging near source regions, while the second bump corresponds to τ = ~1 day, which reflects the aging over rural areas and in the middle troposphere. The third small bump is around τ = 100 h, mainly representing the very slow aging in remote regions (e.g., Polar regions and the upper troposphere)."

4. *"For the figures that show ratios (such as Figure 10), I suggest to try a blue-to-red color scale, where blue represents values <1 and red represents values > 1."*

Response: Points are well taken. We have modified the original Figs. 10-13 by using a blue-to-red color scale (blue: <1; red: >1). Please see the new Figs. 11-14 in the track-changed manuscript.

5. *"Page 32797, line 26: The authors attribute the remaining model-observation discrepancy to BC emissions, wet scavenging and meteorological fields. Is it possible to be more specific which of these three factors contributes most?"*

Response: We thank the reviewer for this constructive comment. Previous studies (Molod et al., 2012; Bond et al., 2013; Q. Wang et al., 2014) suggest that BC emissions, wet scavenging, and meteorological fields could contribute to the model-observation discrepancy during HIPPO BC measurements. However, the present study mainly focuses on the improvement of BC aging process in the model, and thus to quantify the contribution of emissions, wet scavenging, and meteorological fields is beyond the scope of this study. But we note that it is critically important to figure out the contribution of each factor to discrepancies between simulated and observed BC concentration, which can lead to understanding model weakness and further improving BC simulations, subjects required further investigations. We have included corresponding discussions in Page 24, Lines 569-571 as follows:
"We note that it is important to quantify the contribution of these factors to the model-observation discrepancies, which will be investigated in our future study."

6. *"Page 32801, line 1: How do you reconcile the GMD from the HIPPO observations with the range used for the model simulations?"*

Response: Based on the observed size range of fresh BC near combustion sources (Bond et al., 2006), we used 30–90 nm for the geometric mean diameter (GMD) of hydrophobic BC *number* size distribution. However, the observed BC GMD of ~180 nm from HIPPO is for *mass* size distribution. The GMD of mass size distribution converted from its number size distribution (30–90 nm) is about 60–180 nm, close to the observed values from HIPPO. Furthermore, most BC particles observed from HIPPO are highly aged, particularly over the remote Pacific so that they could be larger than BC particles near emission sources (used in this study) due to coagulation and aggregation processes during atmospheric aging. To clarify, we have modified the original statement in Page 28, Lines 685-689 as follows:

"We note that the observationally constrained accumulation mode BC mass size distributions for HIPPO have a geometric mean diameter of ~180 nm (Schwarz et al., 2010), which is the upper bound value used in this study for the geometric mean diameter of mass size distribution (~ 60–180 nm) converted from that of number size distribution (30–90 nm)."

We have also added "number size distribution" in Table 1 (footnote) and in Sects. 2.2.1.1 and 2.2.4 to clarify between mass and number size distributions..

Minor Comments:

1. *"Page 32780, Line 2: The parameterization also accounts for photochemical aging, please add this to the abstract."*

Response: Points are well taken. We have added the chemical aging in the abstract as follows:

"We develop and examine a microphysics-based black carbon (BC) aerosol aging scheme that accounts for condensation, coagulation, and heterogeneous chemical oxidation processes in a global 3-D chemical transport model … and compute the microphysical BC aging rate (excluding chemical oxidation aging) explicitly from the condensation ... and coagulation … The chemical oxidation aging is tested in the sensitivity simulation. …"

2. *"Page 32781, line 12: Even at emission BC is actually frequently mixed with OC (see for example Willis et al., Atmos. Chem. Phys. Discuss., 15, 33555-33582, 2015, and references therein.)"*

Response: We thank the reviewer for this comment. We have incorporated additional discussions and suggested references in Page 4, Lines 60-62 as follows:

"Recent studies showed that BC can also be frequently mixed with organics even at emission under specific conditions (Willis et al., 2015 and references therein)."